

REMARKS

Concurrently with the filing of a Request for Continued Examination (RCE) Transmittal, and as the necessary Submission therefor, Applicants have further amended their claims in order to further clarify the definition of various aspects of the present invention. Specifically, Applicants have amended claims 13, 14, 15, 20 and 24 to recite that the negative electrode comprises graphite powder having a crystal structure, with the crystal structure including a hexagonal crystal structure and/or a rhombohedral crystal structure, these claims defining a hexagonal crystal fraction and/or rhombohedral crystal fraction of the crystal structure of the graphite powder, and also defining a particle size of the graphite powder.

Similarly, each of claims 17-19 has been amended to recite that the carbon material is composed of graphite powder having crystal structure, with a particle size of the graphite powder being defined and with a fraction of hexagonal crystal structure of the crystal structure of the graphite powder being defined. Claims 32 and 33 have been amended to recite that the crystal structure of the graphite powder includes at least a fraction having hexagonal crystal structure.

Applicants are adding new claims 34-38 to the application. These new claims, dependent respectively on claims 13, 14, 17, 20 and 24, recite that the graphite powder has substantially completely the crystal structure.

Applicants respectfully submit that all of the claims presented for consideration by the Examiner patentably distinguish over the teachings of the reference applied by the Examiner in rejecting claims in the Office Action mailed April 21, 2004, that is, the teachings of U.S. Patent No. 5,340,670 to Takami, et al, under the provisions of 35 U.S.C. §102 and 35 U.S.C. §103.

It is respectfully submitted that this reference as applied by the Examiner would have neither taught nor would have suggested such a non-aqueous secondary battery, or such electrodes for a non-aqueous battery having, inter alia, a negative electrode, wherein the negative electrode includes graphite powder having a particle size equal to or smaller than 100 μm , and a hexagonal fraction, of the crystal structure of the graphite powder, being in a range of at least 80% by weight (see claims 14, 15 and 24), or having a rhombohedral fraction, of the crystal structure of the graphite powder, being in the range of 0-20% by weight (see claims 13, 20 and 24), or having both a rhombohedral crystal structure of at most 20% by weight and a fraction of hexagonal crystal structure of at least 80% by weight (see claim 24; note also claim 21).

Furthermore, it is respectfully submitted that this applied reference would have neither disclosed nor would have suggested such electrode for a non-aqueous secondary battery having, inter alia, a negative electrode, as in the present claims, and wherein an active material of the negative electrode is carbon material, this carbon material being composed of natural graphite powder having a crystal structure and having a particle size equal to or smaller than 100 μm , and wherein a fraction of hexagonal crystal structure in the crystal structure of the graphite powder is at least 80% by weight (see claims 17-19), and, moreover, wherein the graphite crystal powder has a deintercalating capacity for lithium of at least 320 mAh/g (see claim 19).

In addition, it is respectfully submitted that this applied reference would have neither taught nor would have suggested such non-aqueous secondary battery or such electrode as in the present claims, wherein the graphite powder has substantially completely the crystal structure (see claims 34-38).

The invention as claimed in the above-identified application is directed to a non-aqueous secondary battery using a negative electrode made of graphite powder, as well as to such negative electrode of graphite powder. The present invention has use, for example, in connection with a lithium battery, and provides such battery which can have a high energy density and a long life, and which has excellent intercalation capacity (that is, an increased capacity of the battery).

Carbon material has been proposed as negative electrode active material in place of lithium metal, in lithium batteries, in which charged and discharged reactions involving lithium ion intercalation into the carbon material and deintercalation from the carbon material occur. However, in previously proposed batteries (e.g., lithium secondary batteries) using carbon material as active material for the negative electrode, a large capacity cannot be obtained; and, moreover, preferable performances of rapid charging and discharging are not obtained. Note the paragraph bridging pages 2 and 3 of Applicants' specification.

Against this background, Applicants provide a negative electrode using carbon powder as active material, which active material has good charging and discharging characteristics, while having a large capacity (large intercalation capacity). Applicants have found that by using graphite powder having a crystal structure (in particular, wherein the graphite powder has substantially completely a crystal structure), with this crystal structure being at least 80% hexagonal crystal structure and/or at most 20% rhombohedral crystal structure, for the overall structure of the graphite powder (which graphite powder, e.g., has laminated graphite layers); and, in particular, by limiting the amount of rhombohedral crystal structure of the graphite powder, and increasing the hexagonal crystal structure thereof, various advantages are achieved. In particular, by reducing the crystal structure of the

graphite powder which is rhombohedral and increasing hexagonal crystal structure thereof, intercalation capacity of the negative electrode is increased. That is, the number of storage sites for lithium anodes in the negative electrode is increased.

According to the present invention, the graphite powder has a crystal structure (specifically, has substantially completely the crystal structure), with the hexagonal crystal structure, of this crystal structure of this graphite powder, being increased, to have the specified minimum hexagonal crystal structure (or, correspondingly, having the specified maximum rhombohedral crystal structure), due to an orderliness and regularity of the hexagonal-net-plane layers. That is, it is respectfully submitted that the fundamental hexagonal crystal structure, of the crystal structure of the graphite powder, according to the present invention, is achieved based upon regularity and orderliness of the lamination. In accordance with the present invention, the existing ratio of hexagonal crystal structure, of the crystal structure of the graphite powder as a whole, is specified at a high level, in order to achieve the increased capacity of the battery as described in Applicants' disclosure.

In comparison, and as will be discussed infra, Takami et al. discloses crystallites and structure with hexagonal-net-plane layers, which are laminated; however, this reference is silent with respect to orderliness and regularity of the laminated hexagonal-net-plane layers. It is emphasized that Takami, et al discloses crystallites, not crystals of the carbon material. In connection with differences therebetween, attention is respectfully directed to the definition of "crystal" and of "crystallite" on page 327 of Hawley's Condensed Chemical Dictionary (12th ed. 1993). As can be appreciated therefrom a crystallite is that portion of a crystal whose constituent atoms, ions, or molecules form a perfect lattice, without strains or other imperfections, and is usually microscopic, (while crystals may be quite large).

Taking the disclosure of Takami, et al as a whole, including the disclosure therein of “appropriate displacements, twists, and angles” of the planar layers to one another (note, e.g., column 7, lines 21-40 of the Takami, et al), it is respectfully submitted that Takami, et al would have neither disclosed nor would have suggested, and in fact would have taught away from, the graphite powder having a crystal structure, and with specified fractions of the crystal structure being hexagonal and/or rhombohedral, as in the present claims, and advantages thereof.

To emphasize, in accordance with the present invention, graphite powder, of a relatively small size, is used as material for the negative electrode, and a focus thereof is on crystallinity of the graphite powder. Applicants have found that by using graphite powder having a crystal structure, in particular, having substantially completely the crystal structure, and having a relatively large amount of this crystal structure being hexagonal crystal structure (and having regularity and orderliness between laminated layers), and, in particular, with this crystal structure of the graphite powder having a specific ratio of hexagonal crystal structure to rhombohedral crystal structure, improvements in capacity are achieved. That is, according to the present invention the graphite powder has a graphite crystal structure, and Applicants limit the amount of rhombohedral structure and increase amount of hexagonal structure, of this crystal structure of the powder of the negative electrodes, with orderliness and regularity between the laminate layers of the graphite, so as to achieve unexpectedly better results of increased capacity, as seen in Applicants’ disclosure.

Takami, et al discloses a negative electrode carbonaceous material suitable for a lithium secondary battery, as well as a battery using this negative electrode carbonaceous material. According to Takami, et al, the negative electrode contains

a carbonaceous material which has an exothermic peak at 700° C or more when measured by a differential thermal analysis, and an intensity ratio P_{101}/P_{100} of a (101) diffraction peak P_{101} to a (100) diffraction peak P_{100} of a graphite structure obtained by X-ray diffraction analysis, of 0.7-2.2; and absorbs and desorbs lithium ions. See column 3, lines 1-8. Other negative electrodes disclosed in Takami, et al, are described at column 3, lines 16-25, 38-44, 56-60 and 66-68; as well in column 4, lines 9-13, 17 and 18.

Note also column 20, lines 25-56; column 21, lines 3-22; column 22, lines 1-19 and 37-62; and column 23, lines 10-21, for other disclosures in connection with negative electrodes of the secondary battery of Takami, et al. These disclosures all refer to the carbonaceous material having a graphite structure that exhibits a property of allowing reversible absorption and desorption of lithium ions to and from hexagonal-net-plane layers in the graphite structure. Note also, for example, example 1 in column 24 and particularly lines 23-29 thereof, disclosing that the carbonaceous material used was a graphitized carbon powder with an average particle size of 25 μm , which was distributed at a ratio of 90 vol.% within the range of 1-50 μm in a particle size distribution, and in which the ratio of particles with a particle size of 0.5 μm or less was 0 vol. % in the particle size distribution. Note also, inter alia, example 2 and the discussion in connection therewith at column 25, lines 20-26; and example 3 and the discussion in connection therewith at column 25, lines 52-59, describing particle size of the carbonaceous material.

Attention is particularly directed to column 7, lines 29-40, of Takami, et al, emphasizing that the carbonaceous material having the recited intensity ratio has appropriate displacements, twists and angles of the hexagonal-net-plane layers so that lithium ions diffuse more easily between the hexagonal-net-plane layers; and

note that also column 14, lines 21-39, disclosing that the carbonaceous material has a mean length L_a of a crystallite in the a-axis direction of graphite structure. This patent again refers to crystallite structure, e.g., at column 22, lines 37-62.

It is respectfully submitted that the disclosure in Takami, et al, having, e.g., crystallites and having hexagonal-net-plane layers, would have neither disclosed nor would have suggested the presently claimed structure, including the graphite powder having the crystal structure (in particular, having substantially completely the crystal structure), with the recited particle size of the graphite powder and with specified hexagonal and/or rhombohedral fraction of the crystal structure of the graphite powder, and advantages thereof, as described in the foregoing.

The contention by the Examiner that the language in column 14, lines 30-43, of Takami, et al (that the carbonaceous material is of a crystallite structure in a hexagonal-net-plane layer) meets Applicants' claimed subject matter of an anode hexagonal crystal structure, is respectfully traversed, particularly insofar as applicable to the claims as presently amended. That is, the claims now expressly recite that the graphite powder has crystal structure, (more specifically, in claims 24-38, recite that the graphite powder has substantially completely the crystal structure). Thus, it is respectfully submitted that the claims recite the graphite powder, and that such graphite powder has crystal structure, not to individual layers of the laminate thereof and/or crystallites of the carbonaceous material. Properly construing the language of the present claims, it is respectfully submitted that Takami, et al would have neither disclosed nor would have suggested the graphite powder having crystal structure (more particularly, having substantially completely the crystal structure), with hexagonal-rhombohedral fractions thereof and with the powder having the recited particle size, and advantages thereof as discussed previously.

In the Advisory Action mailed August 24, 2004, the Examiner has noted that Applicants have not specifically claimed a negative electrode having a hexagonal crystal structure as a whole, and that Applicants have not specifically recited regularity and orderliness of the hexagonal crystal structure of the negative electrode. The present claims recite that the graphite powder has a crystal structure, and, more specifically, in claims 34-38 recite that the graphite powder has substantially completely the crystal structure. In light thereof, and again noting differences between crystallites and crystals, as seen by the enclosed definition from Hawley's Condensed Chemical Dictionary, it is respectfully submitted that Takami, et al would have neither taught nor would have suggested the presently claimed structure, including structure of the graphite powder of the negative electrode, and advantages thereof.

In the first full paragraph on page 3 of the Office Action mailed April 21, 2004, the Examiner has pointed out that Takami, et al, teaches "a crystallite of the graphite anode material", and apparently from this has concluded that Takami, et al teaches a hexagonal crystal structure of the anode. However, as can be appreciated from the enclosed dictionary definition referred to previously a crystallite structure would have neither disclosed nor would have suggested the crystal structure of the graphite powder as in the present invention. Particularly noting the requirement of appropriate displacements, twists and angles of the stacked hexagonal-net-plane layers, in Takami, et al, it is respectfully submitted that this reference would have taught away from the crystal structure of the graphite powder as in the present invention, and advantages thereof.

Reference by the Examiner in this first full paragraph on page 3 of the Office Action mailed April 21, 2004, to a consultation with an expert Examiner in the carbon

field within the U.S. Patent and Trademark Office, is noted. If the Examiner intends continued reliance on this "consultation", it is respectfully submitted that the Examiner must obtain a Declaration/Affidavit of the "Expert Examiner". See 37 C.F.R. § 1.104(d)(2).

It is respectfully submitted that the "displacements, twists and angles" in Takami, et al, disclosed as being necessary for lithium ions to diffuse more easily between the hexagonal-net-plane layers, are contrary to the crystal structure of the graphite powder according to the present invention; and, in particular, are contrary to the orderly and regular lamination in this crystal structure. Based on the difference in regularity of the laminated structure and advantages achieved by the present structure the graphite powder used according to the present invention differs at least in this regard from the structure of Takami, et al. Noting that Takami, et al, provides no description as to regularity and orderliness of the hexagonal-net-plane layers, and in fact requires appropriate displacement, twists and angles, this reference would have neither taught nor would have suggested the presently claimed invention, including wherein the graphite powder has a crystal structure, more specifically substantially completely the crystal structure, and hexagonal and/or rhombohedral fraction thereof. To be specific, it is respectfully submitted that the disclosure of graphite having hexagonal-net-plane structure, without more, would have neither taught nor would have suggested the graphite powder having crystal structure, as in the present claims; and, especially in light of the requirement in Takami, et al of displacements, twists and angles, Takami, et al would have taught away from the crystal structure of the graphite powder according to the present invention.

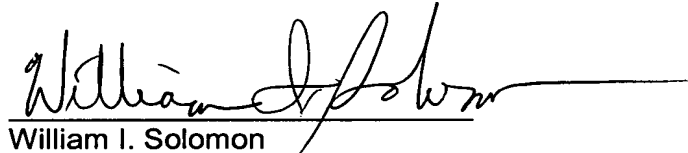
The contention by the Examiner in the last paragraph on page 3 of the Office Action mailed April 21, 2004, is noted. Contrary to the conclusion by the Examiner, it

is respectfully submitted that Takami, et al, does not set forth "the same carbon crystal material", as a whole, as in the present claims; and, accordingly, the contention by the Examiner that it is inherent that the material of Takami, et al would have the same capacity that Applicants have claimed, is respectfully traversed.

In view of the foregoing comments and amendments, and further in view of the concurrently filed RCE Transmittal, entry of the present amendments, and reconsideration and allowance of all claims presently in the application, are respectfully requested.

To the extent necessary, Applicants petition for an extension of time under 37 CFR §1.136. Please charge any shortage in the fees due in connection with the filing of this paper, including extension of time fees, to Deposit Account No. 01-2135 (Case No. 503.34465VV4) and please credit any excess fees to such deposit account.

Respectfully submitted,

A handwritten signature in black ink, appearing to read "William I. Solomon", with a long horizontal line extending to the right.

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WIS/kmh

Attachments

BEST AVAILABLE COPY

Hawley's
Condensed Chemical
Dictionary

TWELFTH EDITION

Revised by
Richard J. Lewis, Sr.



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New York

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CIP

Derivation: Synthetic product is made by fusing NaF and aluminum fluoride.

Use: Electrolyte in the reduction of alumina to aluminum; ceramics; insecticide; binder for abrasives; electric insulation; explosives; polishes.

"Cryovac" [Grace]. TM for a light, shrink-film, transparent packaging material based on polyvinylidene chloride. Used especially for meats and other perishables.

cryptands. See cavitands.

cryptocyanine. (1,1'-diethyl-4,4'-carbocyanine iodide). $C_{22}H_{25}N_2I$.

Properties: Solid, mp 250-5.

Use: Organic dye, soluble, used as a chemical shutter in laser operation.

See also cyanine dye.

cryptostegia rubber. Rubber from leaves of *Cryptostegia grandiflora* and *C. madagascariensis*.

cryptoxanthin. (provitamin A; hydroxy- β -carotene). $C_{40}H_{56}O$. A carotenoid pigment with vitamin A activity.

Properties: Garnet-red prisms with metallic luster; mp 170C; soluble in chloroform, benzene, and pyridine; slightly soluble in alcohol and methanol.

Occurrence: In many plants, egg yolk, butter, blood serum. Can be made synthetically.

Use: Nutrition, medicine.

crystal. The normal form of the solid state of matter. Crystals have characteristic shapes and cleavage planes due to the arrangement of their atoms, ions, or molecules, which form a definite pattern called a lattice. Crystals may be face-centered, body-centered, cubic, orthorhombic, monoclinic, prismatic, etc. They have flat surfaces, sharp edges, and a definite angle between a given pair of surfaces. The form of a crystal is called its "habit." Among the most important features of a crystal are its optical properties, chief of which is its index of refraction, i.e., the extent to which a beam of light is slowed on passing through the crystal. With respect to light transmission, a crystal may be isotropic or anisotropic. Anisotropic crystals can polarize light (see also optical isomer, optical rotation). Crystals also have electrical and magnetic properties now being used in computers and other electronic devices. Crystals are almost always imperfect and contain impurities (atoms of other elements). These are utilized in semiconductors. For methods of growing crystals, see nucleation.

Single crystals are used in masers, lasers, semiconductors, miniaturized components, and

computer memory systems, and as "whiskers." Many metals are now available in large single crystal form and such natural crystals as ruby, garnet, sapphire, etc., are used in these applications.

See also crystallization, nucleation, liquid crystal, hole, vacancy.

crystal face. The recurring characteristic surface of a crystal or a plane parallel to it.

crystal face, indices. Reciprocals of intercepts of a crystal plane on reference axes based on a chosen system of coordinates.

crystal-growth step. A ledge on the surface of a crystal, one or more lattice spacings high, where crystal growth occurs.

crystalline rocks. Igneous or metamorphic rock.

crystal, liquid. See liquid crystal.

crystallite. That portion of a crystal whose constituent atoms, ions, or molecules form a perfect lattice, without strains or other imperfections. Single crystals may be quite large, but crystallites are usually microscopic.

See also crystal.

crystallization. The phenomenon of crystal formation by nucleation and accretion. The freezing of water into ice is one of the commonest examples of crystallization in nature. Industrially, it is used as a means of purifying materials by evaporation and solidification. The sugar of commerce is made in this way. Similarly, salt cake is derived from crystallization of natural brines (e.g., Searles Lake). Nucleated crystallization is also used to form polycrystalline ceramic structures.

See also crystal.

crystallogram. A photograph of an x-ray diffraction pattern of a crystal.

crystallographic systems. (crystal systems). A categorization of crystals according to their degree of symmetry. They are cubic, hexagonal, orthorhombic, tetragonal, monoclinic, and triclinic. The cubic form has the highest symmetry.

crystallography. The study of the crystal formation of solids, including x-ray determination of lattice structures, crystal habit, and the shape, form, and defects of crystals. When applied to metals, this science is called metallography.

crystals of Venus. See copper acetate.

crystal systems. See crystallographic systems.